TACOMA SMELTER PLUME PROJECT
EXTENDED FOOTPRINT STUDY

July 2005

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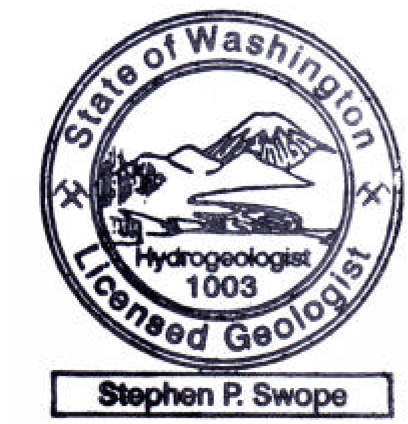
GLOSSARY

Aliquot	Portion of a subdivided sample analyzed for a given analyte
Boring	Vertical section of soil which may be divided into depth intervals for sampling.
Depth Interval	Depth at which soil is sampled from a boring. Up to four depth intervals were sampled within a boring: 0-2, 2-6, 6-2, and 12-18 inches
Distribution	All values within one mile of a given location.
Location	The geographic center of all borings within 300 feet of each other.
mg/kg	Unit of measure of soil concentrations by mass: milligrams of analyte per kilo-gram of sample.
Polygon	An area in a map over which the median and 90^{th} percentile is calculated.
Sample	A single collection of soil from a specific location and depth interval.

SIGNATURE

This report, and Pacific Groundwater Group's work contributing to this report, were reviewed by the undersigned and approved for release. This work was performed, our findings obtained, and this report prepared, using generally accepted hydrogeologic practices used at this time and in this vicinity, for exclusive application to the Tacoma Smelter Plume Extended Footprint Study, and for the exclusive use of the Washington State Department of Ecology and the Health Departments of King, Thurston, Pierce, and Kitsap Counties. This is in lieu of other warranties, express or implied.



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EXECUTIVE SUMMARY

The Asarco Smelter, located in Ruston, Washington, north of Tacoma, operated from 1890 to 1986 primarily as a lead and copper smelter. The smelter (referred to as the Tacoma Smelter for the purposes of this report) was also a major domestic producer of arsenic, with annual production rates on the order of 10,000 tons. The Tacoma Smelter and surrounding areas were included as part of the designated Commencement Bay Superfund site in the early 1980s. In the early 1990s, EPA issued Records of Decision for cleanup actions at the smelter property and in the surrounding Ruston/North Tacoma areas within about one mile of the smelter (Glass, 2004). Cleanup activities at both of these Operable Units are continuing. Near-surface soil contamination was identified as the primary concern in the residential neighborhoods.

A zone of elevated soil metals concentrations extends downwind for several hundred square miles from the Tacoma Smelter and has been defined as the Tacoma Smelter Plume (TSP). Within the TSP, shallow soil metals concentrations have been detected above Model Toxics Control Act (MTCA) Method A cleanup levels. From a health perspective, the primary constituents of concern associated with the TSP are arsenic (Method A cleanup level 20 mg/kg) and lead (Method A cleanup level 250 mg/kg).

The TSP characteristics have been documented in three Initial Footprint studies. These studies include the Vashon-Maury Island Soil Study, the King County Mainland Study, and the Pierce County Footprint Study (see Section 1.1 for full references). The studies were completed between 2000 and 2004. Subsequent to the Initial Footprint Studies, the Department of Ecology and local Health Departments undertook the Extended Footprint Study to address data gaps in Pierce and King Counties and collect samples in Thurston and Kitsap Counties.

The purpose of this report is to summarize and evaluate analytical data collected during the Initial and Extended Footprint Study phases of the Tacoma Smelter Plume Project including the magnitude and large scale spatial patterns of elevated soil levels of arsenic and lead caused by air deposition from the Asarco Smelter. This was accomplished through development of a contouring methodology and use of distance versus concentration scatter plots. In addition, lead/arsenic ratios and contaminant depth profiles were evaluated.

The Initial and Extended Footprint Studies resulted in the collection of 4175 samples from 851 locations and 1928 borings. Samples were collected from 0-2 and 2-6 inches below ground surface for all borings, and deeper samples from 6-12 and 12-18 inches were collected from some borings. The samples were analyzed for arsenic, lead, and for the Vashon-Maury Island Study, cadmium. Conclusions supported by the analysis of the data include the following:

Concentrations: Arsenic was detected in over 99 percent of samples collected and concentrations ranged over four orders of magnitude from 0.48 mg/kg to 1100 mg/kg. These values are compared to background concentrations of 7 mg/kg and the MTCA A cleanup level of 20 mg/kg. Arsenic was detected above the 7 mg/kg background level in 93 percent of samples collected. Arsenic was detected above the 20 mg/kg cleanup level in 55 percent of samples collected. Exceedences of the cleanup level were found in all counties at all depth intervals. Lead was detected in over 97 percent of samples collected and ranged over three orders of magnitude from 1 to 6700 mg/kg. Lead was detected above the MTCA A cleanup level of 250 mg/kg in 17 percent of samples collected. Cadmium was detected in 45.7 percent of the samples collected and concentrations ranged from undetected (at 0.5 mg/kg) to 15 mg/kg. Cadmium was detected above the MTCA A cleanup level of 2 mg/kg in 19 percent of samples collected, and not detected above the MTCA B cleanup level of 80 mg/kg.

Spatial Distribution: Arsenic concentrations are generally highest near the smelter and down the dominant wind directions towards Maury Island and southern Vashon Island, and toward Tacoma and University Place. Concentrations generally decrease with distance from the former smelter site. However, areas of similar concentrations of arsenic are not continuous, i.e. areas of 20-100 mg/kg appear intermixed with areas of 0-20 mg/kg, most notably in the north east of the study area. In general, the edge of the elevated concentration zone was identified and most of the study zone is bounded by unaffected areas. However, areas above the 20 mg/kg level appeared at the edge of the study area in some locations for maps of all depth profiles and percentiles, suggesting the boundary of the 20 mg/kg zone may not have been identified in all cases. This conclusion is consistent with distance versus arsenic concentration plots. In many cases, however, the polygon value is based on a single sample. Further sampling may change this conclusion.

Arsenic/Lead Ratios: Lead is measured at higher concentrations than arsenic in most samples. Non-parametric Spearman rank correlations resulted in correlation coefficients ranging from 0.55 to 0.79. This suggests a statistically significant relationship exists between lead and arsenic. Average lead/arsenic ratios ranged from 1:2.7 in Thurston County to 1:7.1 in Pierce County.

Depth Profiles: Depth-concentration relationships in soil samples generally show higher concentrations of both arsenic and lead in the 0-2 inch shallow samples than the 2-6 inch deep samples. Only 105 of 1,717 samples show higher arsenic concentrations in the 2-6 inch samples than the 0-2 inch samples.

1.0 INTRODUCTION

The Asarco Smelter, located in Ruston, Washington, north of Tacoma, operated from 1890 to 1986 primarily as a lead and copper smelter. The smelter (referred to as the Tacoma Smelter for the purposes of this study) was also a major domestic producer of arsenic, with annual production rates on the order of 10,000 tons. It specialized in the smelting of complex (e.g., high arsenic) ores. Resulting air emissions from the production process included both discharges from the tall smoke stack and fugitive emissions of dust or other contaminated soils from processes at the ground level.

The Tacoma Smelter property itself and surrounding areas were included as part of the Commencement Bay Superfund site in the early 1980s. After smelter closure in 1986, the U.S. Environmental Protection Agency (EPA) reviewed available information and completed extensive investigations of the smelter property and nearby residential neighborhoods. In the early 1990s, EPA issued Records of Decision for cleanup actions at the smelter property and in the surrounding Ruston/North Tacoma areas within about one mile of the smelter. Cleanup activities at these Operable Units are continuing. Near-surface soil contamination was identified as the primary concern in the Ruston/North Tacoma residential neighborhoods.

A zone of elevated soil metals concentrations extends downwind over several hundred square miles from the Tacoma Smelter and has been defined as the Tacoma Smelter Plume (TSP). Within the TSP, shallow soil metals concentrations have been detected above Model Toxics Control Act (MTCA) Method A cleanup levels. From a health perspective, the primary constituents of concern associated with the TSP are arsenic (Method A cleanup level 20 mg/kg) and lead (Method A cleanup level 250 mg/kg).

1.1 PREVIOUS INVESTIGATIONS

In 1999, Ecology and the local health departments in King and Pierce counties began a systematic and phased approach to soil investigations of smelter impacted areas outside of the Ruston/North Tacoma and Asarco Superfund sites. Two sequential study phases, termed "footprint" and "child-use area" sampling, were carried out in a series of defined geographic subregions.

Footprint sampling, always performed first, focused on relatively undisturbed forested areas to develop information on the likely highest levels of soil contaminant concentrations. The studies were guided by a conceptual model of aerial deposition from the smelter stack influenced by wind direction and intensity, and topographic features. The conceptual model predicted higher concentrations in the surface soil down predominant wind directions, higher soil concentrations closer to the smelter, and higher soil concentrations in undisturbed soils. Disturbance during development activities was assumed to dilute the surface soil concentrations.

The initial footprint studies confirmed the conceptual model. In undisturbed soils, arsenic, lead, and other smelter-related contaminants were found to accumulate primarily in the uppermost soils (approximately top foot) resulting in comparatively simple contaminant depth profiles.

Child use area sampling followed footprint sampling, and was conducted in geographic areas identified as having potential for the highest levels of contamination. Child-use area sampling focused on developed areas such as schools, parks, and childcare facilities to develop information on soil contamination in locations where exposures to young children are most likely. The soil disturbance that accompanies property development may diminish the maximum soil contaminant concentrations that would otherwise be present, because of mixing, dilution, placement of fill, and removal of near-surface soils. Contaminant depth profiles thus may be altered substantially. For the most part, child-use area sampling focused on the top 6 inches as representative of soil subject to child exposures under existing conditions (i.e., absent further site development activities that could re-expose deeper and potentially more contaminated soils). The results of previous studies in this series were used to help design subsequent studies. Footprint study results, in particular, were used to identify zones within which child-use properties would be considered for sampling.

The current magnitude and large-scale spatial patterns of soil contamination downwind from the former Tacoma Smelter have been documented in three footprint reports. These reports include the following:

- Tacoma Smelter Plume Site, Pierce County Footprint Study: Soil Arsenic and Lead Contamination in Western Pierce County, Final Report. (Glass, 2004)
- Tacoma Smelter Plume Site, King County Mainland Soil Study Final Report. (Glass, 2002)
- Final Report, Vashon-Maury Island Soil Study. (Seattle and King County Public Health, and Glass, G., 2000)

For all studies, the intent was to focus primarily on undeveloped properties to avoid the effects of dilution and disturbance from development activities. The Vashon-Maury Island Soil study focused only on undisturbed properties due to the nature of the available sampling sites. The King County Mainland Study focused on forested, undisturbed properties between Interstate-90 to the north, the Pierce County line to the south, the Cascade foothills to the east, and Puget Sound to the west. The Pierce County Footprint Study sampled disturbed and undisturbed properties west and north of Interstate-5. Disturbed properties were included in the Pierce County Study because few undisturbed properties were available for sampling. For the most part, the disturbed properties were older residential properties where major disturbance would have occurred decades ago while the smelter was still operating, and thus the air emission impact would still be present in the surface soil. The areas covered by each of these studies is presented in Figure 1.

The Tacoma Smelter Plume studies have been designed and performed through the cooperative efforts of Ecology and local health departments. The studies have been funded through Site Hazard Assessment grants from Ecology to the health departments. For one of the child-use area investigations, Ecology directly procured a contractor to perform the sampling; in all other cases, local health department staff collected the samples.

1.2 OBJECTIVES OF EXTENDED FOOTPRINT STUDY

At completion of the Initial Footprint studies, the full extent of soil contamination had not been identified. The Extended Footprint Study was initiated to identify and fill in data gaps in King and Pierce counties, and extend sampling into Kitsap and Thurston counties in order to determine the extent of the contamination. The overall general objective of the Extended Footprint Study is the same as the Initial Footprint Studies: to provide a better understanding of the extent of TSP contamination.

The specific objectives of the Extended Footprint Study are:

- **Primary Objective:** Refine the concentration pattern of the TSP in the extended footprint study area, especially near the estimated 20 mg/kg arsenic concentration zone, with a high degree of confidence (acceptable level of map stability).
- **Secondary Objective:** Enhance map resolution within the existing area of the currently defined TSP in high sensitivity or low resolution areas.

1.3 REPORT PURPOSE AND OBJECTIVES

This report summarizes data collected as part of the Extended Footprint Study as well as the three previous initial footprint studies discussed in Section 1.1. The purpose of this Extended Footprint Study Report is to summarize and evaluate analytical data collected during the Initial Footprint Studies and Extended Footprint Study phases of the Tacoma Smelter Plume Project, including a description of the magnitude and large scale spatial patterns of elevated concentrations of arsenic and lead in soil caused by air deposition from the Asarco Smelter. Specific objectives of this report include:

- Combine and summarize all footprint data generated from the Tacoma Smelter Plume Project
- Assess the spatial correlation between all arsenic data and the relationship of arsenic concentration to distance and direction from the Asarco Smelter.
- Identify the following four zones of arsenic concentration: 0-20 mg/kg, 21-100 mg/kg, 101-200 mg/kg, and 201-1,050 mg/kg.
- Assess lead/arsenic ratios from the Extended Footprint Study (EFS) data and previous footprint studies
- Evaluate depth profiles from the EFS data and earlier studies

2.0 STUDY DESIGN

Study designs for the initial and extended footprint studies included both study area definition and sample allocation principles. Discussions of both principles are presented below in separate sections. The study designs for the initial footprint studies and Extended Footprint Study are briefly summarized below. The scope of actual sampling is then presented. References for the Footprint Study Designs are:

- Sampling Design for the Tacoma Smelter Plume 2003-2005 Extended Footprint Study (Landau Associates, 2003)
- Sampling Design for Tacoma Smelter Plume Site Pierce County "Footprint" Study (Glass, 2002)
- Sampling Design for Mainland Phase I Study: Further Evaluation of Soil Contamination, King County Mainland Areas, Tacoma Smelter Plume Site (Glass, 2001)
- Sampling Design for Vashon-Maury Island Soils Work Group and Seattle-King County Health Department (Glass, 1999)

2.1 TERMINOLOGY

For clarity, a number of terms related to sampling and sample groupings have been defined for all footprint studies as follows: a *location* is a specific geographic place which may include several borings or samples distributed within the immediate area (300 foot radius), the coordinates of the location were calculated using the centroid of the borings at the location; a *boring* is a vertical hole in the ground from which soil is extracted to a specified depth, and may be the source of multiple samples; a *sample* is a single unit of soil collected at a specific depth interval used for analytical purposes. Figure 2 demonstrates how these groupings are displayed on maps presented in this study.

2.2 STUDY AREA DEFINITION

The overall footprint study area included northeastern Thurston County, along the Puget Sound coastline in eastern Kitsap County, western Pierce County to south and east of I-5, and throughout much of King County from the foothill of the Cascades west to Puget Sound. The study areas for all footprint studies are presented in Figure 1. The study areas for each footprint study (as described in the respective study designs) are:

- **Vashon-Maury Island Soil Study** – the study area was confined to the island's undisturbed (forested) properties.
- **King County Mainland Study** – the study area was defined as forested, undisturbed properties between Interstate-90 to the north, the Pierce County line to the south, the Cascade foothills to the east, and Puget Sound to the west.
- **Pierce County Initial Footprint Study** – the study area was defined as disturbed and undisturbed properties west and north of Interstate-5. Disturbed properties were included in the Pierce County Initial Footprint Study because few undisturbed properties were available for sampling in the more urban portions of the study area. For the most part, the disturbed properties were older residential properties where major disturbance would have occurred decades ago while the smelter was still operating, and thus air emission impact would likely still be present in the surface soil.
- **Extended Footprint Study -** the study area was defined to include the likely extent of smelter impacts, up to and beyond the estimated zone of 20 mg/kg arsenic soil concentration. This zone was estimated based on previous initial footprint study results and included samples from all four counties. The study area was further modified based on other factors such as:
  - Wind channeling
  - Land surface elevation
  - Water-shore air turbulence
  - Land surface slope aspect
  - Puget Sound/land interface

2.3 SAMPLE ALLOCATION

For each footprint study, samples were collected from sample locations usually on a grid, and from 1 to 4 soil borings per sample location. Sample depths varied between studies. The sample allocation is summarized as follows:

- **Vashon-Maury Island Soil Study** - three sampling zones were defined based on wind sector and proximity to the smelter. Sampling grid size (defining density of candidate areas to be sampled) and sampling density (number of borings per area) were specified by zone. A higher sampling density (3 borings per area) was implemented in Zones I and III on about 20% of properties to characterize the scale of concentration variability. Where multiple borings were collected, the distances between borings were between 50 and 300 feet apart. Two depth intervals were sampled at each boring (0 to 2 inch and 2 to 6 inch). Characteristics of the three sampling zones were:
    - Zone I - South Vashon and Maury Island: 1,500 ft grid; 1 boring per location
    - Zone II – Mainland, South King County: Opportunistic (no grid); 3 borings per location
    - Zone III – North Vashon Island: 2,000 ft grid; 1 boring per location
- **King County Mainland Study -** because of the lack of undisturbed properties, an opportunistic sampling design was used that did not rely on a sampling grid. Multiple borings were sampled at each location with distances between borings set at 100 feet and 30 feet. Three depth intervals (0 to 2 inches, 2 to 6 inches and 6 to 12 inches) were sampled at each boring with a fourth depth interval (12 to 18 inches) sampled at about 1/3 of the borings.
- **Pierce County Initial Footprint Study** – this study was based on a more complex sampling design. Six sampling zones were defined depending on disturbance and proximity to the smelter. Developed sampling zones (D1, D2 and D3) included both undisturbed and disturbed properties. However, because developed zones had limited availability of undisturbed properties, extensive disturbed property sampling was undertaken. Undeveloped sampling zones (U1, U2 and U3) contained extensive tracts of undisturbed properties. Therefore, sampling included undisturbed properties exclusively. Two depth intervals were to be sampled (0-2 inches, and 2-6 inches) with 25% to 40% of locations having a third depth interval (6-12 inches). Sampling grids were defined for each sample zone.
    - D1: 2000 ft grid; 4 borings per location
    - D2: 3250 ft grid; 4 borings per location
    - D3: 4500 ft grid; 4 borings per location
    - U1: 3000 ft grid; 3 borings per location
    - U2: 6000 ft grid; 2 borings per location
    - U3: 8000 ft grid; 1 boring per location
- **Extended Footprint Study** – like the Pierce County Initial Footprint study, the extended footprint study includes developed and undeveloped sampling zones. The grid spacing for the undeveloped zone was 8000 feet, with 2 borings per location. For the developed zone, the grid spacing was set at 20,000 feet, but 4 sampling locations were to be selected from within the grid. This was to effectively reduce the grid size to 10,000 feet. Four borings were to be sampled at each location if it was a disturbed

property. For consistency with the initial footprint studies, all borings were to be sampled at 2 depth intervals (0-2 inches, and 2-6 inches).

2.4 SCOPE OF SAMPLING

Seven studies have been completed to date: the three previous footprint studies discussed in Section 1.1 and the four studies completed as part of the Extended Footprint study:

- King County Extended Footprint
- Kitsap County Extended Footprint
- Pierce County Extended Footprint
- Thurston County Extended Footprint

The areas sampled during each study are presented in Figure 1. Table 1 presents an accounting of the number of locations, borings, and samples from each study. Three studies have been completed in King County and include a total of 1776 samples from 421 locations. Samples from Kitsap County were only collected as part of the extended footprint study and include 212 samples from 53 locations. Two studies have been completed in Pierce County resulting in 1982 samples from 277 locations. Samples from Thurston County were only collected as part of the extended footprint study and resulted in 204 samples from 100 locations. The combined data set from all seven footprint studies includes results for 851 properties and 4175 samples.

Samples were collected from four different depths: 0-2, 2-6, 6-12, and 12-18 inches below ground surface. Samples from the 0-2 and 2-6 inch depth intervals were collected for all studies. Samples from the 6-12 inch depth interval were collected during the Pierce County Initial Footprint Study and the King County Mainland Study. Samples from the 12-18 inch depth interval were collected only during the King County Mainland Study.

All samples collected were analyzed for arsenic and lead. Arsenic was analyzed by the Graphite Furnace Atomic Absorption (GFAA) EPA Method SW7060 in the King County Vashon-Maury Island study, and the King County Mainland Study. Arsenic was analyzed by Inductively Coupled Plasma Mass Spectrometry (ICP-MS) for all other studies. Lead was analyzed by Inductively Coupled Plasma Atomic Emission Spectrometer (ICP-AES) EPA Method SW6010. A subset of samples collected from the King County Vashon-Maury Island study was also analyzed for cadmium. Cadmium was analyzed by GFAA using Method SW7131.

Duplicates were collected as part of all studies. The percentage of duplicated samples ranged from 3.7 to 8.2 percent but was generally around 5 percent. Table 2 presents a summary of the number of duplicate samples per study as well as other Quality Assurance (QA) information.

3.0 DATA QUALITY

All footprint studies included a Quality Assurance Project Plan (QAPP) which outlines quality control samples to be collected, and criteria to be met for evaluating the quality of the data. The Initial Footprint studies had full data validation by an independent lab against the data quality objectives of the respective

QAPPs. The results of that data validation are summarized below. The Extended Footprint Study data were not reviewed by an independent lab, as previous data validation supported high quality data from the labs, and resources were limited.

3.1 DATA VALIDATION OF INITIAL FOOTPRINT STUDIES – SUMMARY

Data validation reports were written for the Vashon-Maury Island, King County Mainland, and Pierce County Initial Footprint Studies. No data were rejected and all data were found to be of sufficient quality to perform the analysis.

3.1.1 Vashon-Maury Island Soil Study

An independent data validation review was performed on soil data collected as part of the Vashon-Maury Island Soil Study by Public Health-Seattle & King County. That data validation review followed the procedures established in EPA's Contract Laboratory Program National Functional Guidelines for Inorganic Data Review. The data validation found that the completeness, comparability, and representativeness characteristics of the final data set met the objectives for evaluations of contamination patterns in the defined study area. The field QC results suggested some deficiencies in field protocols, and minor possible effects with data precision and accuracy, but they were judged not to have significant effects on the overall quality of the reported laboratory soils data. No data were rejected for use in data evaluations based on the field QC results. A summary of PHSKC's independent data validation findings is included in the Final Report, Vashon-Maury Island Soil Study (Seattle and King County Public Health, and Glass, G. 2000).

3.1.2 King County Mainland Study

An independent data validation of all arsenic and lead results was performed by EcoChem, Inc. of Seattle, Washington following the procedures established in EPA's National Functional Guidelines for Inorganic Data Review. EcoChem's data validation review did not result in any recommendations for corrective actions at the analytical laboratory during the study and no data were rejected.

3.1.3 Pierce County Initial Footprint Study

An independent data validation of all arsenic and lead results was performed by EcoChem, Inc. of Seattle, Washington following the procedures established in EPA's National Functional Guidelines for Inorganic Data Review. Approximately 10 percent of the sample delivery groups (SDGs) received full validation (Level IV); the remaining 90 percent of SDGs received summary validation (Level III). No data were rejected and the completeness for analytical results was thus 100 percent. EcoChem assigned an "estimated" data quality flag (J flag) to a small proportion of the results, indicating the potential for lesser accuracy or precision in those results compared to unflagged values. All of the J-flagged results were used, as quantified by STL, in data evaluations. EcoChem's data validation reviews did not result in any recommendations for corrective actions at the analytical laboratory during the initial footprint study.

3.2 REVIEW OF DATA QUALITY

For this report, two types of data review were performed on all footprint data - an assessment of data integrity and a comparison of field duplicate values to calculate relative percent differences (RPDs) between the duplicates and primary samples.

Data from all footprint studies were initially downloaded from the Washington State Department of Ecology Environmental Information Management System (EIM). Because multiple data sets were combined from the seven sampling projects, a review of data integrity was performed to assess whether all data collected during the studies were included in the database. Missing values were added to the database and data flags were corrected as necessary. Corrections were made to the EIM data before proceeding with the analysis.

Relative percent differences (RPDs) were used as a measure of the precision of the analytical data – a low RPD reflects high precision. The RPD is calculated between a sample and a duplicate of that sample that is submitted to the same laboratory. The RPD is calculated by dividing the difference between two sample concentrations by the average of the sample concentrations. The Quality Assurance Project Plans set an RPD target of 50 percent or less.

RPDs were calculated for 428 (4.9 percent) of the 8683 aliquots collected as part of all TSP studies. In this report, the term *aliquot* is used to denote the portion of a subdivided sample analyzed for a given analyte. Table 2 presents RPDs broken down by analyte and county as well as the percent of duplicates over the 50 percent target presented in the Quality Assurance Project Plan (QAPP). RPDs were calculated for arsenic, lead, and, for the King County Vashon-Maury Island study, cadmium. Nineteen (four percent) of the 428 duplicates did not meet the 50 percent RPD target stipulated in the QAPP. Nine of those failing were from the King County Vashon-Maury Island study.

Studies with the highest percentage of RPDs above the target of 50 percent and the highest average RPD for all analytes include the King County Vashon-Maury Island Footprint Study (9 percent of RPDs above the target, 27 percent average RPD) and the King County Mainland Footprint Study (4 percent of RPDs above the target, 9.1 percent average RPD). The lowest average RPDs were achieved in the King County Extended Footprint Study (5.8 percent), Pierce County Extended Footprint Study (5.9 percent), and Pierce County Initial Footprint Study (8.2 percent). The differences in RPDs between counties are likely attributed to the laboratories used by each county.

4.0 MAPPING

Previous to this report, data results for all footprint studies were mapped by plotting the maximum concentration detected at a location. While this form of mapping presents the worst-case condition, it does not take into account all of the data and its variability. A primary objective of this report was to identify spatial zones of arsenic concentrations that reflect all of the data. As described in Section 5, a greater percentage of arsenic samples than lead samples exceeded the MTCA cleanup level. Thus mapping was focused on arsenic as representing the extent of the plume contamination. Several methods were considered for developing the maps. A key criterion for method selection was the minimization of data assumptions and the number of parameters estimated.

Global methods of mapping such as Kriging and polynomial surfaces were rejected because of the complexity of the data. Plots of the data distributions (Figures 8 through 23) showed that the concentrations varied strongly with stack distance, wind direction and surface topography. A global model attempting to incorporate the distribution patterns would be overly complex and require estimating many parameters.

Local methods of mapping use only the concentration values from nearby locations to estimate the concentration at a target point. Gridded methods define a grid of target cells and interpolate values for each cell from the values of nearby cells containing sample data. Selection of grid density and interpolation method influence the resultant contours.

Local statistical methods use data from each sample location and its neighbors to specify a distribution of data values. The mean or median and scale (e.g. standard deviation) of the distribution estimate the value and variability of the estimated concentrations. A parametric distribution will estimate the mean and standard deviation by assuming the data are described by a statistical distribution such as the lognormal. The estimated value depends on the choice of distribution. A non-parametric estimate uses the empirical distribution of the data to estimate the median and an upper percentile.

4.1 MAPPING METHODOLOGY

The mapping methodology included dividing the study area into polygons and assigning statistically based concentrations to the polygons. Concentrations were assigned using a non-parametric method. Non-parametric methods do not require that the statistical distribution of the data be known. This method is described in detail in the sections below.

Up to four borings were made at a given location (property) and each boring had up to four different sample depths. Each sample was analyzed for arsenic and lead (and for the Maury-Vashon Island study, also for cadmium). Section 2.1 and Figure 2 present an explanation of the terminology used in this study. To best represent the concentration of an analyte at each location the values from all samples at the same depth within 300 feet of the location were averaged.

The local distribution of values was obtained by pooling the values from the nearest ten locations within one mile to calculate the statistics applied to the corresponding polygon. One mile was selected as the radius which balanced the need to include sufficient points to create a distribution, and the need to include values that were most representative of the location. In cases where fewer than ten locations were within a mile, the statistics were calculated with the available locations. Therefore, statistics were not calculated from values only within the polygon but from the polygon and the ten nearest neighboring polygons.

A distribution was created for each location by combining the ten nearest arsenic concentrations within one mile of the location. Distributions were made from all 0-2 inch depth samples and the combination of the 0-2 and 2-6 inch depth samples. The 0-2 inch depth distributions give an indication of surface concentrations; the combined 0-2 and 2-6 inch depth distributions give an indication of total concentrations within the top 6 inches of the soil. Distributions include between 1 and 10 values depending on the number of locations within one mile of the initial location. For each location, a median and 90^{th} percentile was calculated from the distribution. Either the median or 90^{th} percentile (depending on the map created) were assigned to an area around the location. In cases where no other locations were within a mile, the median and 90th percentile were estimated from the one available value. The method used to calculate the 90^{th} percentile is presented in Appendix A. The area around each location was assigned by constructing Thiessen polygons (see Section 4.3).

4.2 ADJUSTMENTS FOR DIFFERENT NUMBERS OF BORINGS PER LOCATION

At each location, many samples were collected within short distances of each other because multiple borings were drilled at a given location. A location is defined as a specific geographic place which may include several borings (which in turn may contain several samples per boring). A radius of 300 feet was used because it was the intended maximum distance between borings at a location, so the radius should capture all location specific borings. In practice, some borings were made at a distance of greater than 300 feet from each other on a single property. These borings were also included.

All samples from a location were arithmetically averaged before being combined into distributions (used to calculate median and 90th percentile values) in order to evenly weight locations within distributions. If samples were not averaged and all samples were given the same weight, locations with more samples would tend to dominate the subsequently calculated distribution. Duplicates were not included in the averages as stipulated in the QAPP (TPCHD, 1993) and the detection limit was used for non-detect samples.

4.3 THIESSEN POLYGONS

Theissen polygons were used to define the areas to which the median and 90th percentile concentration values were assigned (Thiessen and Alter, 1911). The polygons were constructed by connecting sample locations with line segments, adding perpendiculars to those line segments at their midpoints, and then extending those perpendiculars until they intersected. Finally, the original connecting line segments are removed, leaving irregularly-shaped polygons containing the original locations (Okabe et al. 1992). Figure 2 depicts the Theissen polygons with corresponding line segments. The line segments are called a Triangulated Irregular Network or TIN. Each polygon has the unique property that any point within the polygon is closer to that polygon's location than to the location of any other polygon.

The size of the polygon reflects the sampling density and thus the degree to which an assigned concentration likely reflects arsenic concentration throughout the polygon. Smaller cells reflect higher sampling density. Because the cells are small, samples collected closer to the location are used to develop the statistical distribution and which results in higher confidence in the statistics. Figure 3 presents the number of samples used to create the distributions from which the median and 90th percentiles were calculated. Generally, sampling density is greatest in high concentration areas near the smelter and Vashon-Maury Island.

At the edges of a series of polygons, the outermost polygons are not bounded by other points and extend out infinitely. A system must be applied to terminate the outermost polygons. In this case, the boundary line was drawn such that it was always perpendicular to a polygon boundary. The result was that a boundary of a polygon can only have a vertex on a sample point or at the midpoint between two sample points. This conservative approach minimized extrapolation of the analytical data outside the study area that can not be supported by the data.

4.4 MEDIAN AND 90TH PERCENTILE

The statistics calculated at each location included the median and 90th percentile arsenic concentration. Maps of the median and 90th percentile arsenic concentrations for 0-2 and 0-6 inch depth profiles are presented in Figures 4 through 7. These statistics were calculated using non-parametric methods (described below) and therefore do not assume that the data follow a specific distribution (such as normal or log-normal). Each polygon was colored based on the statistics calculated for each associated location.

The statistics at each location are based on the value at the location and up to ten nearest neighbors (adjacent locations within a mile). Using only the nearest neighbors allows the statistical distribution to track the spatial distribution of concentration values.

The median value is a measure of central tendency; similar to the mean except it is the middle value, above and below which lies an equal number of values. Therefore, the maps depicting median values (Figures 6 and 7) present a reasonably likely value for a given polygon. For a given location, if the samples accurately represent the total area, then the actual value of a sample collected within a polygon has an equal chance of being above the median or below it.

Maps depicting the 90^{th} percentile are presented to indicate higher concentration values that might be expected in a polygon. The 90^{th} percentile value is greater than or equal to 90 percent of the values within the distribution. The 90^{th} percentile was used as opposed to the 95^{th} or 99^{th} because of the generally small distribution size and the skewed nature of the data. The method used to calculate the 90^{th} percentile is presented in Appendix A.

5.0 RESULTS

The following sections present a description of the results including a spatial analysis and correlation of arsenic concentrations, analysis of lead/arsenic ratios, and depth profiles of arsenic and lead. Comparisons are made to several values.

For arsenic:
- 7 mg/kg = natural background (Ecology, 1994)
- 20 mg/kg = MTCA A cleanup level, protection of human health
- 100 mg/kg = interim action level for schools, childcares
- 200 mg/kg = interim action level for parks, camps

For lead:
- 250 mg/kg = MTCA A cleanup level, protection of human health

For cadmium:
- 2 mg/kg = MTCA A cleanup level, protection of groundwater
- 80 mg/kg = MTCA B cleanup level, protection of human health

5.1 MAGNITUDE OF CONCENTRATIONS

Descriptions of arsenic, lead, and cadmium detections are presented below (cadmium was analyzed only for the Maury/Vashon Island initial footprint study). In addition to other statistics, average concentration data are presented broken out by County and depth interval. As with any data reduction, some of the details of data characteristics (e.g., high variability, relationship of concentration to distance from smelter) are lost using this method of presentation. Data summaries are presented in Tables 3 through 9.

5.1.1 Arsenic

Arsenic was detected in 99.7 percent of samples collected. Table 3 presents a summary of arsenic detections versus non-detections. All of the non-detect samples were collected in King County. This may be a function of sampling and analysis techniques in addition to arsenic distribution.

Table 4 presents a summary of minimum, average, and maximum arsenic concentrations detected by county and depth interval. The maximum arsenic concentration of 1100 mg/kg was detected in Pierce County in the 0-2 inch depth range.

King County had the highest average concentration of 32.9 mg/kg for the 0-2 inch depth interval. The lowest average concentration occurred in Kitsap County with 8.2 mg/kg detected in the 2-6 inch depth interval. Average values are presented here as a measure of risk, since they are weighted by concentration.

Detected concentrations ranged from 0.48 mg/kg to 1100 mg/kg compared to a background concentration of approximately 7 mg/kg (Ecology, 1994). Table 5 presents the number of locations with detections greater than 7 mg/kg. The detection limit for non-detected values was higher than the lowest detected value. Arsenic was detected above the 7 mg/kg background level in 93 percent of samples.

Arsenic was detected above the 20 mg/kg MTCA Cleanup Level in all counties and in all depth ranges. King County had the highest percentage (65.8 percent) of locations with arsenic detections above 20 mg/kg. Table 5 presents the number of locations with detections greater than 20 mg/kg. The lowest rate of detections greater than 20 mg/kg of 24.5 percent occurred in Kitsap County.

5.1.2 Lead

Lead was detected in 97.9 percent of samples collected. Table 6 presents a summary of lead detections versus non-detections. As with arsenic, all of the non-detect samples were collected in King County. Concentrations ranged from 1 to 6700 mg/kg. The detection limit for non-detected values was higher than the lowest detected value. Table 7 presents a summary of minimum, average, and maximum lead concentrations detected by county and depth interval. The maximum lead concentration of 6700 mg/kg was detected in Pierce County in the 2-6 inch depth range. The lowest maximum concentration of 132 mg/kg was detected in Kitsap County in the 2-6 inch depth interval.

The highest average lead concentration of 130.7 mg/kg was detected in the 0-2 inch depth interval in Pierce County. The lowest average lead concentration of 20.3 mg/kg was calculated from the 12-18 inch depth interval in King County.

Lead was detected above the MTCA A Cleanup Level of 250 mg/kg in 145 locations (17 percent). Lead was detected above the Cleanup Level to a depth of 12 inches in King and Pierce Counties and 2 inches in Thurston County. Lead was not detected above the Cleanup Level in Kitsap County.

5.1.3 Cadmium

Cadmium was only analyzed in samples collected in King County as part of the Vashon-Maury Island Study. Table 8 presents a summary of cadmium detections versus non-detections. Cadmium was detected in 45.7 percent of samples collected and concentrations ranged from undetected (at 0.5 mg/kg) to 15 mg/kg. Table 9 presents a summary of minimum, average, and maximum cadmium concentrations detected by depth interval. The maximum concentrations detected were 14 mg/kg in the 0-2 inch depth interval and 15 mg/kg in the 2-6 inch depth interval. Average concentrations were 1.4 mg/kg in the 0-2 inch depth interval and 1.2 mg/kg in the 2-6 inch depth interval.

Cadmium was detected above the MTCA A Cleanup Level of 2 mg/kg in 34 locations (19 percent), and was detected above the MTCA A Cleanup Level to a depth of 12 inches. Unlike arsenic and lead for which the MTCA A cleanup level is based on the protection of human health, the MTCA A cleanup level for cadmium is based on the protection of groundwater. The cadmium cleanup level for protection of human health is the MTCA B cleanup level of 80 mg/kg. Cadmium was not detected above the MTCA B cleanup level. As cadmium was not detected above cleanup levels for protection of human health, it was dropped as an analyte in the subsequent footprint studies.

5.2 ARSENIC CONCENTRATION CONTOURS

Figures 4 through 7 present maps of the median and 90^{th} percentile arsenic concentrations for samples collected from the 0-2 inch and 0-6 inch depth intervals. The 0-6 inch depth interval was calculated by combining values of samples collected from the 0-2 inch and 2-6 inch depth intervals. The 0-2 inch depth maps give an indication of surface contamination and the 0-6 inch depth maps present the whole profile sampled. Similar concentration maps for each county are presented in Appendices B through E. The methods used to prepare the maps are presented in Section 4.0.

Previous to this report, data results for all footprint studies were mapped by plotting the maximum concentration detected at a location. These maps are included in Appendix G. While this form of mapping presents the worst-case condition, it does not take into account all of the data and its variability.

For all maps, the highest concentrations are generally near the smelter, and in the dominant downwind directions on Maury Island, southern Vashon Island, and parts of Pierce County. Concentrations generally decrease with distance from the former smelter site. However, areas of similar concentration are not continuous, i.e. areas of 20-100 mg/kg appear intermixed with areas of 0-20 mg/kg, most notably in the north east of the study area. This may be an artifact of the concentration intervals selected for the maps and the low density of sampling locations. The concentration intervals were selected as: 0-20 mg/kg (below MTCA A); 20-100 mg/kg (between MTCA A and Ecology established interim action level for schools, childcares); 100-200 mg/kg (between interim action level for schools, childcares and interim action level for parks, camps); and >200 mg/kg (above interim action levels).

As discussed in Section 5.5, deeper samples tend to have lower concentrations. Therefore, combining shallow and deep samples should result in lower concentrations than shallow samples alone. However, for the two 90^{th} percentile maps, both the 0-2 and 0-6 inch sample depth maps have a generally similar number of polygons for each concentration range, although the 0-2 inch sample depth map has more 20-100 mg/kg concentration polygons in the northeast and southwest portions of the study area.

In general, the edge of the elevated concentration zone was identified; therefore, most of the study area is bounded by unaffected areas (0-20 mg/kg polygons). The bounding is affected by the percentile used to define the boundary. Also, as indicated in the distance versus concentration scatter plots (Section 5.3), higher concentrations can occur outside a lower concentration zone. The notable exceptions to the 0-20 mg/kg bounds are on the east end of the study site immediately south of Interstate-90 and the area immediately surrounding Fort Lewis/McChord Air Base in the south end of the study area. The area immediately south of Interstate-90 corresponds with the E-W trending topographic escarpment that includes Tiger, Rattlesnake, and Cougar Mountains. This raised escarpment likely results in increased deposition of airborne metals and therefore increases concentrations in this area. This phenomenon may occur elsewhere in unsampled areas along the foothills of the Cascades.

Access was not granted to sample the area immediately surrounding Fort Lewis and McChord Air base. Collection of samples there would likely identify the 20 mg/kg boundary in this area.

Both 90th percentile maps (Figures 4 and 5), show two other study area boundary segments above 20 mg/kg - on the southeast side of the study site, near the King County/Pierce County border; and on the north side of the study site just west of Interstate-5. The 90th percentile, 0-2 inch depth profile map also indicates an area on the southwest end of the study site south of Interstate-5.

5.3 SPATIAL CORRELATION

Scatter plots of distance versus arsenic concentration were used as an additional method to assess arsenic distribution. The plots were constructed along the 16 directions of the wind rose as shown in Figure 3. Data were sorted into 16 “spokes” of the wind rose originating at the Tacoma Smelter. Figures 8 through 23 plot distance from the smelter versus log-scaled arsenic concentration, including all samples, with the exception of duplicate QA/QC analyses. A log-scaled axis was used since the data more closely approximate a lognormal distribution than a normal distribution. The MTCA Method A unrestricted use (20 mg/kg) and TSP Interim Action Trigger level (100 mg/kg) limits for arsenic are included on the plots for reference.

In most of the plots, arsenic concentrations are below 20 mg/kg cleanup level at the furthest extent of sampling. However arsenic concentrations are still at or above the cleanup standards near the study boundary in the following directions:

- **Southwest** – Most of the sample concentrations are below 20 mg/kg and the trend of the maximum concentrations is decreasing (Figure 14).
- **East-Northeast** – As with the southwest direction, most of the samples collected at the edge of the sampling are below 20 mg/kg and the maximum concentrations are decreasing (Figure 21).
- **Northeast** – Most of the sample concentrations are below 20 mg/kg but the trend does not appear to decrease (Figure 22). In fact, over the 30 to 40 mile interval, maximum concentrations appear to be increasing. The frequency of water bodies and topographic relief along this wind direction could account for the protracted elevated concentrations.
- **North-Northeast** – Similar to the southwest and east-northeast plots, maximum arsenic concentrations are at the 20 mg/kg level at the study boundary and appear to be decreasing (Figure 23).

All four of these wind rose directions are in dominant wind directions and land masses from which samples were collected are in close proximity to significant water bodies and topographic rises. These two factors likely affect the distance over which arsenic is distributed.

During the initial Footprint Studies, arsenic concentrations were still above the 20 mg/kg clean up level at the boundary of the study area. Arsenic concentrations were assumed to decay exponentially away from the smelter in the 2002 study. Therefore, the distance to the 20 mg/kg concentration level was estimated by fitting an exponential curve to the upper limit of the data (a straight line in a log-linear plot). The 2002 method is reproduced in Appendix F using data from the Extended Footprint Study to assess its applicability to the few areas identified in Section 5 where arsenic concentration had not decreased below 20 mg/kg. In addition to the newly available data, all data between 0 and 6 inches was used to improve extrapolations.

5.4 LEAD/ARSENIC RATIOS

Lead is measured at higher concentrations than arsenic in most samples. Figures 24 through 27 present arsenic versus lead concentration plots for each county in the study area. Most points plot above the 1:1 line, reflecting higher concentrations of lead than arsenic. Lead concentrations in Pierce County visually show a 'bulge' away from the 1:1 line within the 8-40 mg/kg arsenic range. The maximum frequency of arsenic concentrations lies in this interval, and the 'bulge' is consistent with an increase in the number of lead concentration outliers due to a larger number of lead samples in that region of the plot. The elevated lead concentrations may also reflect urban lead concentrations.

Lead and arsenic do not correlate well using standard correlation methods, resulting in R^2 values between 0.18 and 0.56. Table 10 presents a summary of all lead versus arsenic correlation data.

The nonparametric Spearman rank correlation method was also used to assess the strength of association of lead and arsenic. The Spearman method was selected because it is a nonparametric method and therefore the statistical distribution of the data does not need to be known. The Spearman method assigns a rank to each observation in each group separately, then calculates the sums of the squares of the differences in paired ranks. Spearman coefficients may range from 1 to -1, with 1 indicating perfect correlation and -1 indicating anti-correlation.

The Spearman rank correlation resulted in significantly higher correlation coefficients, ranging from 0.55 to 0.79. All of the Spearman Rank correlations were significant at the 99th percentile. The high Spearman correlation coefficients and significance percentiles suggests a statistically significant relationship exists between lead and arsenic. Average lead/arsenic ratios ranged from 1:2.7 in Thurston County to 1:7.1 in Pierce County.

5.5 DEPTH PROFILES

Soil samples were collected from the 0-2 inch and 2-6 inch depth profiles for all studies. Analysis of these data is presented here. Samples from the 6-12 and 12-18 inch depth profiles were collected from a subset of the studies and analysis of those samples has previously been presented in Glass (2004) and Glass (2002).

Soil samples in the shallow (0-2 inch) samples generally show higher concentrations of both arsenic and lead than deeper (2-6 inch) samples. Figures 28 through 31 plot shallow versus deep soil arsenic concentration in each county. Figures 32 through 35 plot shallow versus deep soil lead concentration in each county. Points plotting below the 1:1 line indicate lower concentrations in the shallow samples relative to the corresponding deeper sample. Values that plot above the 1:1 line indicate higher concentrations in the shallow sample.

Arsenic concentrations are generally higher in the 0-2 inch shallow soil samples. Figure 36 is a histogram of the ratios of arsenic concentrations in the 0-2 versus 2-6 inch depth profiles. Ratios greater than one plotting to the right of the figure indicate that the 0-2 inch depth sample has a higher arsenic concentration than the 2-6 inch depth sample and that concentrations decrease with depth. As indicated by the summations at the top of the figure, 948 pairs have a ratio greater than one indicating the 0-2 inch sample is of higher concentration than the 2-6 inch sample compared to only 105 sample pairs that have a ratio lower than one. These results indicate that arsenic concentrations decrease with depth in the majority of borings.

Lead concentrations are generally higher in the shallow (0-2 inch) samples than the deep (2-6 inch) samples. Figure 37 is a histogram of the ratios of lead concentrations in the 0-2 versus 2-6 inch depth profiles, similar to Figure 36. Ratios greater than one plotting to the right of the figure indicate that the 0-2 inch depth sample has a higher concentration of lead than the 2-6 inch depth sample and that concentrations decrease with depth. As with the arsenic concentrations, the summations at the top of the figure indicate that there are more depth ratios above one (1278) than below one (77). Therefore, lead concentrations also generally decrease with depth.

6.0 CONCLUSIONS

The following conclusions may be drawn from this study and are generally consistent with previous studies:

Concentrations: Arsenic was detected in over 99 percent of samples collected and detected concentrations ranged over four orders of magnitude from 0.48 mg/kg to 1100 mg/kg. These values are compared to background concentrations of seven mg/kg and the MTCA cleanup level of 20 mg/kg. Arsenic was detected above the 7 mg/kg background level in 93 percent of samples collected. Arsenic was detected above the 20 mg/kg cleanup level in 55 percent of samples collected. Exceedences of the cleanup level were found in all counties at all depth intervals. Lead was detected in over 97 percent of samples collected and ranged over three orders of magnitude from 1 to 6700 mg/kg. Cadmium was detected in 45.7 percent of the samples collected and concentrations ranged from undetected (at 0.5 mg/kg) to 15 mg/kg.

Spatial Distribution: Arsenic concentrations are generally highest near the smelter and down the dominant wind rose directions towards Maury Island, Vashon Island, and parts of Pierce County. Concentrations generally decrease with distance from the site. However, areas of similar concentrations of arsenic are not continuous, i.e. areas of 20-100 mg/kg appear intermixed with areas of 0-20 mg/kg, most notably in the north east of the study area. In general, the edge of the elevated concentration zone was identified and most of the study zone is bounded by unaffected areas. However, polygons above the 20 mg/kg level appeared at the edge of the study area in some locations for maps of all depth profiles and percentiles, suggesting the boundary of the 20 mg/kg zone has not been identified in all cases. This conclusion is confirmed by the distance versus concentration plots.

Arsenic/Lead Ratios: Lead is measured at higher concentrations than arsenic in most samples. Non-parametric Spearman rank correlations resulted in correlation coefficients ranging from 0.55 to 0.79. This suggests a statistically significant relationship exists between lead and arsenic. Average lead/arsenic ratios ranged from 1:2.7 in Thurston County to 1:7.1 in Pierce County.

Depth Profiles: Depth-concentration relationships in soil samples generally show higher concentrations of both arsenic and lead in the 0-2 inch shallow samples than the 2-6 inch deep samples. Only 105 of 1,717 samples show higher arsenic concentrations in the 2-6 inch samples than the 0-2 inch samples.

6.1 DATA GAPS

The following types of data gaps have been identified: areas where data could not be collected due to lack of access, areas where the 20 mg/kg boundary has not been reached due to sampling or analysis design, and areas where data density was insufficient.

Two areas could not be sampled due to lack of access, Fort Lewis and McChord Air base. As shown in Figures 1 and 3 through 7, this omission results in a significant hole in the southern portion of the study area. The area bordering the missing area is above the 20 mg/kg boundary and data from the area would likely constrain the boundary in that area.

Definition of the 20 mg/kg arsenic boundary was addressed in two ways in this report. Figures 4 through 7 present maps of the boundary for two different percentiles and depth ranges. Figures 8 through 23 present distance versus concentration plots with maximum concentration lines extrapolated outside of the data range where the range has not dropped below 20 mg/kg. Both methods identified areas where arsenic concentrations had not decreased below the 20 mg/kg cleanup standard.

Data density differs dramatically across the site. Polygon areas range from 22 to 7,943 acres - a difference of more than two orders of magnitude. Generally the size of the polygons increase with decreasing arsenic concentration – polygons are smaller in high concentration areas, as was intended in the sample design. The difference in data densities results in the higher concentration zones – 100 to 200 and 200 to 1050 being well defined but the lower concentration zones are less well defined. Increasing sample density in lower concentration areas would enhance the resolution in these areas.

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